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(54) Title: MULTILAYER ETHYLENE COPOLYMER FILM

(57) Abstract

A multilayer film and a pouch containing a flowable material are disclosed. The multiplayer film is made from two layers of a sealant film with an interposed layer of a polyethylene. The sealant film is made from a material comprising 10 to 100 parts by weight of a copolymer of ethylene and at least one C4-C10 alpha-olefin manufactured in a polymerization process using a single-site polymerization catalyst and from 0 to 90 parts by weight of at least one polymer selected from the group consisting of a linear copolymer of ethylene and at least one C4-C10 alpha-olephin having a density of from 0.900 to 0.930 g/cm³ and a melt index of from 0.3 to 2.0 dg/min, a high-pressure polyethylene having a density of from 0.916 to 0.930 g/cm³ and a melt index of from about 1 to 10 dg/min, and blends thereof. The polyethylene of the interposed layer is a high density polyethylene with a density of at least 0.930 g/cm³ and a melt index of less than 5 dg/min. The pouch is made from the multilayer film and has transversely sealed ends. The pouches are particularly useful in the packaging of flowable materials e.g. milk, especially using a vertical form, fill and seal apparatus.

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MULTILAYER ETHYLENE COPOLYMER FILM

The invention relates to ethylene copolymer film of improved stiffness and clarity, to multilayer ethylene copolymer film, to pouches formed from multilayer ethylene copolymer film and especially to such pouches filled with flowable materials, e.g. liquids. In particular embodiments, the pouches are made on so-called vertical form, fill and seal apparatus.

As used herein, the term "flowable material" does not include gaseous materials, but encompasses materials which are flowable under gravity or may be pumped. Such materials include liquids e.g. milk, water, fruit juice, oil; emulsions e.g. ice cream mix, soft margarine; pastes e.g. meat pastes, peanut butter; preserves e.g. jams, pie fillings, marmalade; jellies; doughs; ground meat e.g. sausage meat; powders e.g. gelatin powders, detergents; granular solids e.g. nuts, sugar; and like materials. The invention described herein is particularly useful for flowable foods e.g. milk. In addition, as used herein "density" is determined by ASTM Procedure D1505-85, "stiffness" is understood to be tensile modulus of elasticity as measured by ASTM Procedure D882-91 (Method A) and "melt index" by ASTM Procedure D1238-90B (Condition E). The "melting point" of a polymer is measured as the peak melting point when performing differential scanning calorimetry (DSC) as described in ASTM Procedure D3417-83 (rev. 88).

Aspects of the invention relate to the packaging of flowable materials e.g. milk, in pouches using for example a so-called vertical form, fill and seal apparatus. Using such an apparatus, a flat web of synthetic thermoplastic film is unwound from a roll and formed into a continuous tube in a tube-forming section, by sealing the longitudinal edges on the film together to form a so-called lap seal or a so-called fin seal. The tube thus formed is pulled vertically downwards to a filling station. The tube is then collapsed across a transverse cross-section of the tube, the position of such cross-section being at a sealing device below the filling station. A transverse heat seal is made, by the sealing device, at the

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collapsed portion of the tube, thus making an air-tight seal across the tube. The material being packaged enters the tube above the transverse heat seal in a continuous or intermittent manner, thereby filling the tube upwardly from the transverse heat seal. The tube is then allowed to drop a predetermined distance usually under the influence of the weight of the material in the tube. The jaws of the sealing device are closed again, thus collapsing the tube at a second transverse section, which may be at, above or below the air/material interface in the tube, depending on the nature of the material being packaged and the mode of operation of the process. The sealing device seals and severs the tube transversely at the second transverse section. The material-filled portion of the tube is now in the form of a pillow shaped pouch. Thus, the sealing device has sealed the top of the filled pouch, sealed the bottom of the next-to-be-formed pouch and separated the filled pouch from the next-to-be-formed pouch, all in one operation.

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One vertical form, fill and seal apparatus of the type described above is a Prepac® IS-6 liquid packaging apparatus.

A sealing device commonly used is a so-called impulse sealer which has a sealing element mounted in sealing jaws and electrically insulated therefrom. In operation, the sealing jaws are closed and an electrical current is caused to flow through a sealing element e.g. a wire, for a fraction of the time that the jaws are closed. The jaws remain closed while the seal forms. Once the sealing jaws are open the synthetic thermoplastic film must be able to provide a transverse seal that supports the weight of the flowable material e.g. liquid, in the next-to-be-formed pouch.

Pouches made from ethylene/butene-1 copolymer films tend to suffer from defective seals i.e. a tendency to have weak transverse end and/or longitudinal seals even though the operating conditions of the impulse sealer have been optimized. Defective seals may lead to the phenomenon known as "leakers", in which the flowable material e.g. milk, may escape from the pouch through pinholes that develop at or close to the seal. It has been estimated that leakers account for about 1-2% of the

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1.3 litre milk pouch production.

U.S. 4,521,437 of W.J. Storms, issued June 04, 1985, discloses the use of pouches of ethylene/octene-1 copolymer film in the packaging of flowable materials; the film disclosed has a stiffness of about 30 000 psi when the polymer of the film has a density of 0.920 g/cm^3 . That patent discloses that pouches made from the ethylene/octene-1 copolymer provide superior leaker performance i.e. a lower percentage of leakers, compared with related films described above that are formed from linear ethylene/butene-1 copolymers. Further improvements in leaker performance may be achieved using film formed from a linear ethylene copolymer manufactured using a single-site polymerization catalyst. However certain films made from the ethylene copolymers manufactured using the single site polymerization catalyst may have lower stiffness than the film of Storm's patent. Lack of stiffness may adversely affect the runnability of the film on a form, fill and seal apparatus and gives poor stand-up properties for pouches in, for instance, a milk pitcher.

It has now been found that the stiffness of film formed from polyethylene manufactured using a single-site polymerization catalyst may be increased in a manner that further improves the formation of pouches.

Drawings

Figure 1 shows leaker rate for a pouch drop test.

Description of Invention

This invention provides for a skin of single-site catalyst films (SSC) and core layer of stiffer polymer such as high density polyethylene (HDPE). HDPE films used by itself, in the packaging industry tends to be hazy when viewed. It is desirable for such HDPE film used in packaging to be clear particularly when used to make pouches for milk to present a desirable looking product for consumption. It has been found that the clarity of films, made with skins (on either side of the core layer) of SSC and core layer of HPDE film, are improved.

The present invention provides for a ethylene co-polymer

film having

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- (a) a sealant film comprising a copolymer of ethylene and at least one C_4 C_{10} alpha-olefin manufactured in a polymerization process using a single site polymerization catalyst, and
- (b) a stiffening layer of polyethylene.

The layer of polyethylene is interposed the sealant film. The sealant film preferably has a density in the range of 0.86 to 0.94 g/cm³ and a melt index of 0.2 to 10 dg/min. and most preferably has a density in the range of 0.88 to 0.93 g/cm³ and a melt index of 0.3 to 5 dg/min.

The sealant film further comprises at least one polymer selected from the group comprising a linear copolymer of ethylene and at least one C_4 - C_{10} alpha-olefin, a high pressure polyethylene and blends thereof. Preferably the sealant film composition comprises 10 to 100 parts by weight of a copolymer of ethylene and at least one C_4 - C_{10} alpha-olefin manufactured in a polymerization process using a single-site polymerization catalyst and from 0 to 90 parts by weight of at least one polymer selected by the group comprising a linear copolymer of ethylene and blends thereof. In particularly preferred embodiments, the composition comprises 20-100 and especially 50-100 parts of the copolymer obtained using the single site catalyst, and conversely 0-80 and 0-50 parts of the other polymers specified above.

The thickness of the sealant film and interposed layer of polyethylene may vary in accordance with this invention. In one embodiment of this invention, for example, the thickness of the interposed layer of polyethylene is approximately one mil (or 25 microns) and the sealant film approximately one mil (or 25 microns) while in another embodiment the thickness of the interposed layer of polyethylene is in the range of 5 to 110 microns with the total thickness of the film being in the range of 40 to 130 microns.

Moreover, other embodiments of the invention may have the thickness of the interposed layer of polyethylene between 5 and 50 microns or between 5 to 20 microns. Furthermore, the melt

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index of the interposed layer of polyethylene in other embodiments may be less than 1 dg/min.

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The present invention also provides a polyethylene film formed from two layers of a sealant film and an interposed layer of polyethylene; said sealant film being made from a composition comprising 10 to 100 parts by weight of a copolymer of ethylene and at least one $C_4\text{-}C_{10}$ alpha-olefin manufactured in a polymerization process using a single-site polymerization catalyst and from 0 to 90 parts by weight of at least one polymer selected from the group consisting of a linear copolymer of ethylene and at least one C4-C10 alphaolefin having a density of from 0.90 to 0.930 g/cm3 and a melt index of from 0.3 to 2.0 dg/min, a high-pressure polyethylene having a density of from 0.916 to 0.930 g/cm3 and a melt index of from about 1 to 10 dg/min, and blends thereof; said interposed layer of polyethylene being of a thickness in the range of 5 to 110 microns and being formed from polyethylene having a density of at least $0.93~\mathrm{g/cm^3}$ and a melt index of less than 5 dg/min; said laminate having a thickness of from 40 to 130 microns.

invention also provides a multilayer present polyethylene film formed from two layers of a sealant film and an interposed layer of polyethylene; said sealant film being made from a composition comprising 10 to 100 parts by weight of a copolymer of ethylene and at least one C_4 - C_{10} alpha-olefin manufactured in a polymerization process using a single-site polymerization catalyst and from 0 to 90 parts by weight of at least one polymer selected from the group consisting of a linear copolymer of ethylene and at least one C4-C10 alphaolefin having a density of from about 0.900 to 0.930 g/cm3 and a melt index of from 0.3 to 10.0 dg/min, a high-pressure polyethylene having a density of from 0.916 to 0.930 g/cm3 and a melt index of from about 1 to 10 dg/min, and blends thereof, said sealant film being selected on the basis of providing pouches with an M_{11} -test value that is lower, at the same film thickness, than that for pouches made with a film of a blend of 85 parts of a linear ethylene/octene-1 copolymer having a

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density of about 0.920 g/cm³ and a melt index of about 0.75 dg/min and 15 parts of a high pressure polyethylene having a density of about 0.918 g/cm³ and a melt index of 8.5 dg/min; said interposed layer of polyethylene being of a thickness in the range of 5 to 110 microns and being formed from polyethylene having a density of at least 0.93 g/cm³ and a melt index of less than 5 dg/min; said laminate having a thickness of from 40 to 130 microns. The M_{11} test is described below.

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The present invention further provides a multilayer polyethylene film formed from a layer of a sealant film and a layer of polyethylene; said sealant film being made from a composition comprising 10 to 100 parts by weight of a copolymer of ethylene and at least one C_4 - C_{10} alpha-olefin manufactured in a polymerization process using a single-site polymerization catalyst and from 0 to 90 parts by weight of at least one polymer selected from the group consisting of a linear copolymer of ethylene and at least one $C_4\text{-}C_{10}$ alpha-olefin having a density of from 0.900 to 0.930 g/cm3 and a melt index of from 0.3 to 2.0 dg/min, a high-pressure polyethylene having a density of from 0.916 to 0.930 g/cm^3 and a melt index of from about 1 to 10 dg/min, and blends thereof; said layer of polyethylene being of a thickness in the range of 5 to 110 microns and being formed from polyethylene having a density of at least 0.93 g/cm³ and a melt index of less than 5 dg/min; said multilayer film having a thickness of from 40 to 130 microns.

The present invention additionally provides a film formed from two layers of a sealant film and an interposed layer of a film having a higher stiffness than the sealant layer, said layer of higher stiffness being formed from a polymer having a melting point of not more than 140°C; said sealant film being made from a composition comprising 10 to 100 parts by weight of a copolymer of ethylene and at least one C_4 - C_{10} alpha-olefin manufactured in a polymerization process using a single-site polymerization catalyst and from 0 to 90 parts by weight of at least one polymer selected from the group consisting of a linear copolymer of ethylene and at least one C_4 - C_{10} alpha-

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olefin having a density of from 0.900 to 0.930 g/cm³ and a melt index of from 0.3 to 2.0 dg/min, a high-pressure polyethylene having a density of from 0.916 to 0.930 g/cm³ and a melt index of from about 1 to 10 dg/min, and blends thereof;

said interposed layer of higher stiffness being of a thickness in the range of 5 to 110 microns and being formed from polyethylene having a density of at least 0.93 g/cm³ and a melt index of less than 5 dg/min; said laminate having a thickness of from 40 to 130 microns.

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The present invention also provides a multilayer film formed from a layer of a sealant film and a layer of a film having a higher stiffness than the sealant layer, said layer of higher stiffness being formed from a polymer having a melting point of not more than 140°C; said sealant film being made from a composition comprising 10 to 100 parts by weight of a copolymer of ethylene and at least one C_4 - C_{10} alpha-olefin manufactured in a polymerization process using a single-site polymerization catalyst and from 0 to 90 parts by weight of at least one polymer selected from the group consisting of a linear copolymer of ethylene and at least one C_4 - C_{10} alpha-olefin having a density of from 0.900 to 0.930 g/cm³ and a melt index of from 0.3 to 2.0 dg/min, a high-pressure polyethylene having a density of from 0.916 to 0.930 g/cm³ and a melt index of from about 1 to 10 dg/min, and blends thereof;

said layer of higher stiffness being of a thickness in the range of 5 to 110 microns;

said multilayer film having a thickness of from 40 to 130 microns.

In preferred embodiments of the multilayer film of the present invention, the film has a stiffness of at least 20 000 psi, and especially at least 25 000 psi.

The present invention also provides a pouch containing a flowable material, said pouch being made from a multilayer film in tubular form and having transversely heat sealed ends, said multilayer polyethylene film being formed from two layers of a sealant film and an interposed layer of polyethylene; said sealant film being made from a composition comprising 10

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to 100 parts by weight of a copolymer of ethylene and at least one C_4 - C_{10} alpha-olefin manufactured in a polymerization process using a single-site polymerization catalyst and from 0 to 90 parts by weight of at least one polymer selected from the group consisting of a linear copolymer of ethylene and at least one C_4 - C_{10} alpha-olefin having a density of from 0.900 to 0.930 g/cm³ and a melt index of from 0.3 to 2.0 dg/min, a high-pressure polyethylene having a density of from 0.916 to 0.930 g/cm³ and a melt index of from about 1 to 10 dg/min, and blends thereof; said interposed layer of polyethylene being of a thickness in the range of 5 to 110 microns and being formed from polyethylene having a density of at least 0.93 g/cm³ and a melt index of less than 5 dg/min; said multilayer film having a thickness of from 40 to 130 microns.

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The present invention also provides a pouch containing a flowable material, said pouch being made from a multilayer film in tubular form and having transversely heat sealed ends, said multilayer polyethylene film being formed from two layers of a sealant film and an interposed layer of polyethylene;

said sealant film being made from a composition comprising 10 to 100 parts by weight of a copolymer of ethylene and at least one $C_4\text{-}C_{10}$ alpha-olefin manufactured in a polymerization process using a single-site polymerization catalyst and from 0 to 90 parts by weight of at least one polymer selected from the group consisting of a linear copolymer of ethylene and at least one C_4 - C_{10} alpha-olefin having a density of from about 0.900 to 0.930 g/cm^3 and a melt index of from 0.3 to 10.0 dg/min, ahigh-pressure polyethylene having a density of from 0.916 to $0.930~{\rm g/cm^3}$ and a melt index of from about 1 to 10 dg/min, and blends thereof, said sealant film being selected on the basis of providing pouches with an M_{11} -test value that is lower, at the same film thickness, than that for pouches made with a film of a blend of 85 parts of a linear ethylene/octene-1 copolymer having a density of about 0.920 g/cm³ and a melt index of about 0.75 dg/min and 15 parts of a high pressure polyethylene having a density of about 0.918 g/cm^3 and a melt index of 8.5 dg/min; said interposed layer of polyethylene being of a thickness in

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the range of 5 to 110 microns and being formed from polyethylene having a density of at least 0.93 g/cm³ and a melt index of less than 5 dg/min; said multilayer film having a thickness of from 40 to 130 microns.

In a preferred embodiment of the laminate and pouches of the present invention, the copolymer made with a single-site catalyst has a density in the range of 0.8-0.93 g/cm³ especially in the range of 0.88 to 0.91 g/cm³; and a melt index of less than 5 dg/min particularly in the range of 0.3 to 2 dg/min, and especially in the range of 0.5 to 1.5 dg/min.

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In a further embodiment, the copolymer made with a singlesite catalyst is an ethylene/butene-1 copolymer, an ethylene/hexene-1 copolymer, ethylene/octene-1 copolymer or an ethylene/hexene-1/butene-1 copolymer.

The present invention further provides a process for making pouches filled with a flowable material, using a vertical form, fill and seal apparatus, in which process each pouch is made from a flat web of film by forming a tubular film therefrom with a longitudinal seal and subsequently flattening the tubular film at a first position and transversely heat sealing said tubular film at the flattened position, filling the tubular film with a predetermined quantity of flowable material above said first position, flattening the tubular film above the predetermined quantity of flowable material at a second position and transversely heat sealing said tubular film at the second position, the improvement comprising making the pouches from a flat web of film made from a multilayer film formed from two layers of a sealant film and an interposed layer of polyethylene;

said sealant film being made from a composition comprising 10 to 100 parts by weight of a copolymer of ethylene and at least one C_4 - C_{10} alpha-olefin manufactured in a polymerization process using a single-site polymerization catalyst and from 0 to 90 parts by weight of at least one polymer selected from the group consisting of a linear copolymer of ethylene and at least one C_4 - C_{10} alpha-olefin having a density of from 0.900 to 0.930 g/cm³ and a melt index of from 0.3 to 2.0 dg/min, a high-pressure

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polyethylene having a density of from 0.916 to 0.930 g/cm³ and a melt index of from about 1 to 10 dg/min, and blends thereof; said interposed layer of polyethylene being of a thickness in the range of 5 to 110 microns and being formed from polyethylene having a density of at least 0.93 g/cm³ and a melt index of less than 5 dg/min; said multilayer film having a thickness of from 50 to 100 microns.

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The present invention also provides, in a process for making pouches filled with a flowable material, using a vertical form, fill and seal apparatus, in which process each pouch is made from a flat web of film by forming a tubular film therefrom with a longitudinal seal and subsequently flattening the tubular film at a first position and transversely heat sealing said tubular film at the flattened position, filling the tubular film with a predetermined quantity of flowable material above said first position, flattening the tubular film above the predetermined quantity of flowable material at a second position and transversely heat sealing said tubular film at the second position, the improvement comprising making the pouches from a flat web of film made from a composite formed from two layers of a sealant film and an interposed layer of polyethylene;

said sealant film being made from a composition comprising 10 to 100 parts by weight of a copolymer of ethylene and at least one C_4 - C_{10} alpha-olefin manufactured in a polymerization process using a single-site polymerization catalyst and from 0 to 90 parts by weight of at least one polymer selected from the group consisting of a linear copolymer of ethylene and at least one C_4 - C_{10} alpha-olefin having a density of from about 0.900 to 0.930 g/cm³ and a melt index of from 0.3 to 10.0 dg/min, a high-pressure polyethylene having a density of from 0.916 to 0.930 g/cm³ and a melt index of from about 1 to 10 dg/min, and blends thereof, said sealant film being selected on the basis of providing pouches with an M_{11} -test value that is lower, at the same film thickness, than that for pouches made with a film of a blend of 85 parts of a linear ethylene/octene-1 copolymer having a density of about 0.920 g/cm³ and a melt index of about

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0.75 dg/min and 15 parts of a high pressure polyethylene having a density of about 0.918 g/cm³ and a melt index of 8.5 dg/min.

Although melt index ranges are specified herein, it is understood that the polymers have melt indices typical of film-grade polymers.

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Although the invention may be a multilayer film of a sealant film and another layer, especially polyethylene, and the related pouches and process described herein, the invention will be particularly described with reference to a multilayer or composite of two layers of sealant film and an interposed layer of polyethylene film. The latter multilayer films are preferred because of the ability to form a lap seal as well as a fin seal and the substantial absence of curl in the laminate. In addition, although the non-sealant layer may be a layer of higher stiffness

formed from a polymer having a melting point of not more than 140°C, that layer will be particularly described herein with reference to a layer of polyethylene.

The multilayer film is made, in part, from a composition comprising 10 to 100 parts by weight of a copolymer of ethylene and at least one C_4-C_{10} alpha-olefin manufactured in a polymerization process using a single-site polymerization catalyst and from 0 to 90 parts by weight of at least one polymer selected from the group consisting of a linear copolymer of ethylene and at least one C4-C10 alpha-olefin having a density of from about 0.900 to 0.930 g/cm3 and a melt index of from 0.3 to 2.0 dg/min, a high-pressure polyethylene having a density of from 0.916 to 0.924 g/cm3 and a melt index of from about 1 to 10 dg/min, and blends thereof. embodiments, the composition comprises 20-100 and especially 50-100 parts of the copolymer obtained using the single site catalyst, and conversely 0-80 and 0-50 parts of the other polymer specified above.

The polyethylene of the interlayer may be a so-called high pressure polyethylene or a so-called linear polyethylene, and blends thereof. In embodiments, the polyethylene is 0-100% by weight of a high density polyethylene, including high molecular

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weight high density polyethylene, preferably 20-100% by weight of such polyethylene and in particular 75-100% by weight of such polyethylene. The remainder of the polyethylene, if any, i.e. 0-80% by weight and especially 0-25% by weight, is a linear polyethylene or a recycled polyethylene obtained from any of a variety of sources provided that the recycled polyethylene meets the appropriate governmental or other regulations for packaging of the material that is to be packaged. The composition used for the interlayer needs to meet the density and melt index values specified herein viz. be a polyethylene having a density of at least 0.93 g/cm³ and a melt index of less than 1 dg/min.

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Processes for the manufacture of so-called high pressure polyethylene from ethylene monomer have been operated commercially for decades. A variety of such polymers are available. The high pressure polyethylene of the sealant layer has a density of 0.916 to 0.930 g/cm³, especially 0.918 to 0.925 g/cm³. In addition, the high pressure polyethylene has a melt index in the range of 1 to 10 dg/min. and especially in the range of 1.5 to 5 dg/min. For the interposed layer, the high pressure polyethylene would have a density and melt index in the range described herein.

Processes for the commercial manufacture of polyethylene have been known for more than thirty years. Such operated at temperatures processes may be above solubilization temperature of the polymer in which event the polymer remains in solution, or the processes may be operated below the solubilization temperature in which event the polymer remains as a slurry in the polymerization solvent. Processes are also known in which the polymerization is conducted in the gas phase in the absence of solvent. Other processes operate at both high temperature and high pressure. The catalysts used are frequently based on titanium and are referred to coordination catalysts; such catalysts may also be described as multi-site catalysts or heterogeneous catalysts. The polymer obtained is linear in nature, as opposed to the branched nature of high pressure polyethylene. Linear low density polyethylene

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is obtained by the co-polymerization of ethylene with at least one C_4 - C_{10} alpha-olefin hydrocarbon comonomer, examples of which include butene-1, hexene-1 and octene-1. The linear low density polyethylene has a density in the range of 0.900 to 0.930 g/cm³, preferably 0.912 to 0.930 g/cm³ and especially in the range of 0.918 to 0.925 g/cm³. In addition the polymer has a melt index in the range of 0.3 to 10.0 dg/min, preferably 0.3 to 2.0 dg/min and especially in the range of 0.5 to 1.5 dg/min.

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The high density polyethylene has a density of at least 0.930 g/cm³, especially in the range of 0.935-0.965 g/cm³. Similarly, the high density polyethylene has a melt index of less than 1 dg/min and especially in the range of 0.01 to 0.3 dg/min; polymer in the lower end of that range may be classified as high molecular weight polyethylenes. The high density polyethylene may be high pressure polyethylene or linear polyethylene.

The ethylene copolymer made with single-site catalyst may be obtained using a variety of polymerization processes of the type described above for the manufacture of linear low density polyethylene e.g. processes that operate in solution, in the gas phase and as a slurry process; references to the use of single site catalysts in polymerization processes is made in Modern Plastics, p.15, May 1993, Plastics Focus Vol. 25, No. 12, June 21, 1993 and in Exxon Chemical Exact Facts, Vol.1, No. 1, February 1993. Such polymers are obtained by the copolymerization of ethylene with at least one C_4 - C_{10} alpha-olefin hydrocarbon comonomer, examples of which include butene-1, hexene-1 and octene-1. The catalyst used is a so-called single-site catalyst, certain of which may also be referred to as metallocene or constrained geometry catalysts.

By the term "single-site catalyst" is meant a metallocene or constrained geometry catalyst. Metallocene catalysts are organometallic co-ordination compounds obtained as a cyclopentadienyl (Cp) derivative of a transition metal or metal halide. The metal is bonded to the Cp ring by electrons moving in orbitals extending above and below the plane of the ring (pi bond). Metallocene catalysts systems are extremely sensitive

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to the geometry of the catalytic site at the transition metal (the "single-site"). Examples of single-site catalysts include Cp₂TICl₂, Cp₂ZrCl₂, Cp₂HfCl₂, (C₅(CH₃)₂)₃TiCl₂, PH₂Me(Ind)₂ZrCl₂, [Me₄Cpsi(Me)₂N(t-Bu)]TiCH₂[o-PhN(Me₂)], Cp₂Fe B(C₂F₂₎₄.

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The polyethylene obtained with the single site catalyst preferably has a density in the range of 0.8 to 0.93 g/cm³, moreover in the range of 0.88 to 0.930 g/cm³, and especially in the range of 0.89 to 0.910 g/cm³. In addition the polymer preferably has a melt index of less than 5 dg/min, particularly in the range of 0.3 to 2.0 dg/min and especially in the range of 0.5 to 1.5 dg/min. Preferred polymers include ethylene/butene-1, ethylene/hexene-1, ethylene/octene-1 and ethylene/hexene-1/butene-1 copolymers.

The multilayer polyethylene film in one embodiment has a thickness of 40 to 130 microns, and especially in the range of 50 to 100 microns. In particular embodiments e.g. for use on a form, fill and seal apparatus, the laminate thickness is preferably in the range of 50 to 70 microns. The sealant layers on opposed sides of the interposed polyethylene layer are preferably of substantially equal thickness, although this is not essential provided that the requirements for the intended end-use are met, to allow the film to be reversed and/or opposing sides to be bonded together as in a tube. The interposed layer has a thickness of 5 to 110 microns, in one embodiment 5 to 50 microns and especially 8 to 16 microns in other embodiments.

The M₁₁-test is as follows: In general terms, the M₁₁-test is a drop test carried out on 1.3 litre water-filled pouches. In the test, pouches containing 1.3 litres of water, at a temperature of 10±5°C, are made from a 32 cm. wide film web, on a Prepac IS-6 vertical form, fill and seal apparatus. The web of film is formed into a continuous tube with a lap seal, said tube having an inside diameter of approximately 9.8 cm. A typical vertical sealing jaw of the Prepac IS-6 apparatus has an approximately 3.5 by 0.3 mm rectangular sealing wire made of Nichrome® alloy, and a transverse sealing jaw with an approximately 1.8 by 0.7 mm sealing wire made of Nichrome®

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alloy. The operation of the vertical and transverse sealing element should be optimized for the particular film type and thickness used. Typically, during vertical sealing of the tube, a 35-50 amp current at 14-24 volts is passed through the sealing wire for about 0.2-0.5 seconds. The force applied by the vertical sealing jaws to the film is about 8 N applied for a dwell time of about 0.9 seconds. The vertical sealing jaw is cooled by water at 13°±6°C. During transverse sealing of the tube, a 35-55 amp current at 10-15 volts is passed through the sealing wire for about 0.2-0.5 seconds. The force applied by the transverse sealing jaws to the film is about 19 N applied for a dwell time of about 0.9 seconds. The transverse sealing jaw is cooled by water at 13°±6°C. Both sealing jaws are covered with 150 μm thick glass fiber tape impregnated with polytetrafluoroethylene resin. With optimizing the sealing operations, it will be recognized that the sealing conditions (e.g. amperage, voltage, dwell time) depend on the gauge and melting characteristics of the film. For example, a $50\mu m$ film would require lower amperage and voltage, as controlled by the rheostat on the apparatus, than would a 75μ m film. Typically such a change in film thickness requires an adjustment of approximately 10% of the rheostat range.

The water-filled pouches containing the cold water, are dropped onto a concrete floor from a height of 335 cm. The pouches are positioned with the longitudinal axis of the tube i.e. pouch, coincident with an imaginary vertical line.

Any pouch from which water flows or weeps after the pouch has been dropped onto the floor, is termed a "leaker". The number of leakers, expressed as a percentage of the total number of pouches dropped is the M_{11} -test value for the particular film being tested. It will be noted that the M_{11} -test value will be affected by the thickness of the film as well as the material from which the film is made.

As noted above, it is the sealant film that is selected upon the basis of a drop test such as the M_{11} -test. It is to be

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understood, therefore, that pouches made with other pouchforming apparatus come within the scope of the present invention.

The blends useful in the present invention may be made by blending the constituents prior to feeding to the hopper of a film extruder, or may be made by blending the constituents at the time of extrusion just prior to remelting in the extruder, or alternatively the constituents may be melt blended in the extruder.

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The ethylene/α-olefin copolymer or blend of ethylene/αolefin copolymer e.g. in pellet form, may be fed into a film
extruder and extruded into film form using known techniques.
One preferred method of manufacturing film is the so-called
blown film process disclosed in Canadian Patent No. 460 963
issued November 8, 1949 to E.D. Fuller. Another preferred
method of manufacturing film uses an internal or external
cooling mandrel in the blown film process, as disclosed for
example in Canadian Patent No. 893 216 issued February 15, 1972
to M. Bunga and C.V. Thomas.

The film, after manufacture, is slit longitudinally into appropriate widths. The width is selected on the basis of the diameter of the tube to be formed on the vertical form, fill and seal apparatus.

The preferred method of manufacture of the multilayer film is by using a blown film coextrusion process, although other methods of manufacture of the film may be used.

It will be understood by those skilled in the art that additives e.g. UV stabilizers, anti-block agents, slip additives, may be added to the polymers from which the pouches of the present invention are made.

The pouches of the present invention may be used in the packaging of flowable materials e.g. liquids, as defined above. In particular, the pouches may be used in the packaging of milk.

The multilayer film of the invention is particularly useful in the formation of pouches, especially using a form, fill and seal apparatus. In particular, the multilayer film

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provides increased stiffness, compared to the monolayer sealant film, increased clarity compared to HDPE film, and further improves efficiencies in the runnability of monolayer sealant films as described herein.

The multilayer film may be used in the formation of pouches, for the packaging of flowable materials. In particular, the multilayer film may be used in the packaging of milk using a form, fill and seal apparatus.

EXAMPLE

10 Introduction

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Films were made on a five-layer blown film coextrusion line. The line consists of five extruders (A through E) feeding on an eight inch diameter circular die. The two "skin layer" extruders A and E were 2.5" single-screw extruders, while the three "core" extruders B, C and D were 2". The line includes an oscillating nip, corona treater, edge guide and a back-to-back winder with slit-in-line, capable of surface-driven or centre-driven winding. The width of the tower nip and winder also allow for blow-up ratios up to about 4:1.

Film Extrusion

Formulations for the skin and core layers were preblended. A control linear low density polyethylene (LLDPE)/low density polyethylene (LDPE) blend, two single-site catalyst polymer formulations, and two high density polyethylenes were chosen:

Control

85% Sclair 11L4B octene LLDPE, density = 0.920 g/cc 15% Nova LF0521A LDPE, density = 0.921 g/cc

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Single Site Catalyst Film (SSC)

SSC #1

52.5% Exact 3028 butene copolymer mPE, 0.900 g/cc density

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40% Exact 4011 butene copolymer mPE, 0.887 g/cc density

densi

3.4% Duex 8876 slip/antiblock concentrate

0.1% MB35102 antiblock concentrate

4.0% SAX 7401 extrusion aid concentrate

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(mPE means metallocene polyethylene)

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SSC #2 92.5% Exact 3033, butene-hexene terpolymer mPE, 0.900 g/cc density 3.4% Duex 8876 slip/antiblock concentrate 0.1% MB35102 antiblock concentrate 4.0% SAX 7401 extrusion aid concentrate Sclair 16A, 0.945 g/cc density HDPE #1

Sclair 19C, 0.957 g/cc density HDPE #2

Film	Layer A (outermost) 1.0 mil	Layer B 0.3 mil	Layer C 0.4 mil	Layer D 0.3 mil	Layer E (innermost) 1.0 mil
Control	85 % Sclair 11L4B 15 % Nova LF0521A	85 % Sclair 11L4B 15 % Nova LF0521 A	85 % Sclair 11L4B 15 % Nova LF0521A	85 % Sclair 11L4B 15 % Nova LF0521A	85 % Sclair 11L4B 15 % Nova LF0521A
Film 1	SSC #1 (Exact 3028-Exact 4011 blend)	85 % Sclair 11L4B 15 % Nova LF0521A	85 % Sclair 11L4B 15 % Nova LF0521A	85 % Sclair 11L4B 15 % Nova LF0521A	SSC #1 (Exact 3028-Exact 4011 blend)
Film 2	55C #1 (Exact 3028-Exact 4011 blend)	Sclair 19C HDPE	Sclair 19C HDPE	Sclair 19C HDPE	SSC #1 (Exact 3028-Exact 4011 blend)
Film 3	SSC #2 (Exact 3033)	85 % Sclair 11L4B 15 % Nova LF0521A	Sclair 16A HDPE	85 % Sclair 11L4B 15 % Nova LF0521A	SSC #2 (Exact 3033)
Film 4	SSC #2 (Exact 3033)	85 % Sclair 11L4B 15 % Nova LF0521A	Sclair 19C HDPE	85 % Sclair 11L4B 15 % Nova LF0521A	SSC #2 (Exact 3033)

All of the above identifiers are either trade-marks or designations available in the trade.

The control film was made at a blow up ration of 2:1. The other films were made at a blow up ratio of 3:1. were wound as a flattened tube and slit to a narrower width. Film Testing

The films were run on the Prepac IS-6 filler to assess runnability, to make pouches for evaluating in a pitcher, and to make pouches for a pouch drop test.

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	<u>Film</u>	Runnability of IS-6 filler	Pouch-in-pitcher
' 5	Control	- good slip - good pouch length	stands up after openingdoesn't fold over edge of pitcher when pouring
10	Film 1	good slipsome drag on forming plates	 stiffness improved relative the SCC film referred to below
15	Film 2	<pre>- pouch length variation, i.e. long- short</pre>	-good stiffness
20	Film 3	-very good slip properties	- very similar to control
	Film 4	-very good slip properties	- very similar to control
30	Previous all-SSC films, such as Exact 3033, Dow Affinity PL1880	- pouch length variation, i.e. pouches get shorter and shorter	- pouch tends to fold over edge of pitcher when pouring, and stays there

For the pouch drop test, films were made into 1.3 litre water-filled pouches. For each pouch drop test, approximately 200 pouches were dropped on end from eleven feet onto a concrete floor. Results of the pouch drop impact test are shown in Figure 1.

40 Analysis

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From the results one can conclude that combining a core of HDPE with skin layers of a single-site or metallocene-catalyst polymers makes a film with improved clarity; adequate stiffness to run well on the existing IS-6; and good pouch drop impact resistance.

A structure such as these with mPE on the skin layers and other polymers in the core would have the advantage of the mPE's lower temperature sealing on the vertical seal.

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Although the preferred embodiments as well as the operation and the use have been specifically described herein, it should be understood that variations in the preferred embodiments could be achieved by a person skilled in the trade without departing from the spirit of the invention as claimed herein.

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CLAIMS

1. A film with a skin of a single-site catalyst polymer and a core of a stiffening polymer.

2. A polyethylene film having:

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- (a) a sealant film comprising a copolymer of ethylene and at least one $C_4\text{-}C_{10}$ alpha-olefin manufactured in a polymerization process using a single-site polymerization catalyst, and
- (b) a stiffening layer of polyethylene.
- 3. The film as claimed in claim 2 wherein said sealant film further comprises at least one polymer selected from the group comprising a linear copolymer of ethylene and at least one C₄-C₁₀ alpha-olefin, a high pressure polyethylene and blends thereof.
- 15 4. The film as claimed in claim 3 wherein said composition of sealant film comprises 10 to 100 parts by weight of said copolymer of ethylene and at least one C_4 - C_{10} alphaolefin manufactured in a polymerization process using a single-site polymerization catalyst.
 - 5. The film as claimed in claim 4 wherein said sealant film comprises 0 to 90 parts by weight of at least one polymer selected from the group consisting of a linear copolymer of ethylene and at least one C_4 - C_{10} alpha-olefin, a high pressure polyethylene and blends thereof.
- 6. The film of claim 5 wherein said sealant film has a density in the range of 0.86 to 0.94 g/cm³ and a melt index in the range 0.20 to 10 dg/min.
 - 7. The film as claimed in claim 6 wherein said stiffening layer of polyethylene has a density of at least $0.93~\text{g/cm}^3$ and a melt index of less than 5~dg/min.
 - 8. The film as claimed in claim 6 wherein said stiffening layer of polyethylene has a thickness of approximately 25 microns and said sealant film has a thickness of approximately 25 microns.
- 9. The film as claimed in claim 7 wherein said stiffening layer of polyethylene is interposed said sealant film and has a thickness in the range of 5 to 110 microns and said film has

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a thickness in the range of 40 to 130 microns.

- 10. A multilayer polyethylene film formed from two layers of a sealant film and an interposed layer of polyethylene; said sealant film being made from a composition comprising 10 to 100 parts by weight of a copolymer of ethylene and at least one $C_4\text{-}C_{10}$ alpha-olefin manufactured in a polymerization process using a single-site polymerization catalyst and from 0 to 90 parts by weight of at least one polymer selected from the group consisting of a linear copolymer of ethylene and at least one C_4 - C_{10} alpha-olefin having a density of from 0.900 to 0.930 g/cm³ and a melt index of from 0.3 to 2.0 dg/min, a high-pressure polyethylene having a density of from 0.916 to 0.930 g/cm³ and a melt index of from about 1 to 10 dg/min, and blends thereof; said interposed layer of polyethylene being of a thickness in the range of 5 to 110 microns and being formed from polyethylene having a density of at least 0.93 g/cm3 and a melt index of less than 5 dg/min; said multilayer film having a thickness of from 40 to 120 microns.
- of a sealant film and an interposed layer of polyethylene; said sealant film being made from a composition comprising 10 to 100 parts by weight of a copolymer of ethylene and at least one C₄-C₁₀ alpha-olefin manufactured in a polymerization process using a single-site polymerization catalyst and from 0 to 90 parts by weight of at least one polymer selected from the group consisting of a linear copolymer of ethylene and at least one C₄-C₁₀ alpha-olefin having a density of from about 0.900 to 0.930 g/cm³ and a melt index of from 0.3 to 10.0 dg/min, a high-pressure polyethylene having a density of
- from 0.916 to 0.930 g/cm³ and a melt index of from about 1 to 10 dg/min, and blends thereof, said sealant film being selected on the basis of providing pouches with an M₁₁-test value that is lower, at the same film thickness, than that for pouches made with a film of a blend of 85 parts of a linear ethylene/octene-1 copolymer having a density of about 0.920 g/cm³ and a melt index of about 0.75 dg/min and 15 parts of a high pressure polyethylene having a density of about 0.918

g/cm³ and a melt index of 8.5 dg/min; said interposed layer of polyethylene being of a thickness in the range of 5 to 110 microns and being formed from polyethylene having a density of at least 0.93 g/cm³ and a melt index of less than 5 dg/min; said multilayer film having a thickness of from 40 to 130 microns.

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- 12. The multilayer film of Claim 10 or Claim 11 in which the copolymer made with a single-site catalyst has a density in the range of 0.88-0.93 g/cm³; and a stress exponent of less than 1.2.
- 13. The multilayer film of Claims 10-12 in which the copolymer made with the single-site catalyst has a melt index of less than 5 dg/min.
- 14. The multilayer film of any one of Claims 10-13 in which the copolymer made with a single-site catalyst is selected from ethylene/butene-1 copolymers, ethylene/hexene-1 copolymers, ethylene/octene-1 copolymers, ethylene/octene-1/butene-1 copolymers and ethylene/hexene-1/butene-1 copolymers.
- 15. The multilayer film of any one of Claims 10-14 in which the sealant film contains 20-100 parts of the copolymer manufactured with a single-site catalyst.
- 16. The multilayer film of any one of Claims 10-14 in which the sealant film contains 50-100 parts of the copolymer manufactured with a single-site catalyst.
- 17. A multilayer polyethylene film formed from a layer of a sealant film and a layer of polyethylene; said sealant film being made from a composition comprising 10 to 100 parts by weight of a copolymer of ethylene and at least one C_4 - C_{10} alphaolefin manufactured in a polymerization process using a single-site polymerization catalyst and from 0 to 90 parts by weight of at least one polymer selected from the group consisting of a linear copolymer of ethylene and at least one C_4 - C_{10} alphaolefin having a density of from 0.900 to 0.930 g/cm and a melt index of from 0.3 to 2.0 dg/min, a high-pressure polyethylene having a density of from 0.916 to 0.930 g/cm³ and a melt index of from about 1 to 10 dg/min, and blends thereof;

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said layer of polyethylene being of a thickness in the range of 5 to 110 microns and being formed from polyethylene having a density of at least 0.93 g/cm³ and a melt index of less than 5 dg/min; said multilayer film having a thickness of from 40 to 130 microns.

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18. A multilayer film formed from two layers of a sealant film and an interposed layer of a film having a higher stiffness than the sealant layer, said layer of higher stiffness being formed from a polymer having a melting point of not more than 140°C;

said sealant film being made from a composition comprising 10 to 100 parts by weight of a copolymer of ethylene and at least one C_4 - C_{10} alpha-olefin manufactured in a polymerization process using a single-site

polymerization catalyst and from 0 to 90 parts by weight of at least one polymer selected from the group consisting of a linear copolymer of ethylene and at least one C₄-C₁₀ alphaolefin having a density of from 0.900 to 0.930 g/cm³ and a melt index of from 0.3 to 2.0 dg/min, a high-pressure polyethylene having a density of from 0.916 to 0.930 g/cm³ and a melt index of from about 1 to 10 dg/min, and blends thereof;

said interposed layer of higher stiffness being of a thickness in the range of 5 to 110 microns and being formed from polyethylene having a density of at least 0.93 g/cm³ and a melt index of less than 5 dg/min; said laminate having a thickness of from 40 to 130 microns.

19. A multilayer film formed from a layer of a sealant film and a layer of a film having a higher stiffness than the sealant layer, said layer of higher stiffness being formed from a polymer having a melting point of not more than 140°C; said sealant film being made from a composition comprising 10 to 100 parts by weight of a copolymer of ethylene and at least one C_4 - C_{10} alpha-olefin manufactured in a polymerization process using a single-site polymerization catalyst and from 0 to 90 parts by weight of at least one polymer selected from the group consisting of a linear copolymer of ethylene and at least one C_4 - C_{10} alpha-olefin having a density of from 0.900 to 0.930 g/cm³

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and a melt index of from 0.3 to 2.0 dg/min, a high-pressure polyethylene having a density of from 0.916 to 0.930 g/cm³ and a melt index of from about 1 to 10 dg/min, and blends thereof; said layer of higher stiffness being of a thickness in the range of 5 to 110 microns;

said multilayer film having a thickness of from 40 to 130 microns.

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- 20. A pouch containing a flowable material, said pouch being made from a multilayer polyethylene film in tubular form and having transversely heat sealed ends, said multilayer polyethylene film being formed from two layers of a sealant film and an interposed layer of polyethylene;
- film and an interposed layer of polyethylene; said sealant film being made from a composition comprising 10 to 100 parts by weight of a copolymer of ethylene and at least one C4-C10 alpha-olefin manufactured in a polymerization process using a single-site polymerization catalyst and from 0 to 90 parts by weight of at least one polymer selected from the group consisting of a linear copolymer of ethylene and at least one C4-C10 alpha-olefin having a density of from 0.900 to 0.930 g/cm³ and a melt index of from 0.3 to 2.0 dg/min, a high-pressure polyethylene having a density of from 0.916 to 0.930 g/cm³ and a melt index of from about 1 to 10 dg/min, and blends thereof; said interposed layer of polyethylene being of a thickness in the range of 5 to 110 microns and being formed from polyethylene having a density of at least 0.93 g/cm³ and a melt index of less than 5 dg/min; said film having a thickness of from 40 to 130 microns.
- 21. A pouch containing a flowable material, said pouch being made from a multilayer polyethylene film in tubular form and having transversely heat sealed ends, said multilayer polyethylene film being formed from two layers of a sealant film and an interposed layer of polyethylene;
- said sealant film being made from a composition comprising 10 to 100 parts by weight of a copolymer of ethylene and at least one C_4 - C_{10} alpha-olefin manufactured in a polymerization process using a single-site polymerization catalyst and from 0 to 90 parts by weight

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of at least one polymer selected from the group consisting of a linear copolymer of ethylene and at least one $C_4\text{-}C_{10}$ alphaolefin having a density of from about 0.900 to 0.930 g/cm³ and a melt index of from 0.3 to 10.0 dg/min, a high-pressure polyethylene having a density of from 0.916 to 0.930 g/cm³ and a melt index of from about 1 to 10 dg/min, and blends thereof, said sealant film being selected on the basis of providing pouches with an M_{11} -test value that is lower, at the same film thickness, than that for pouches made with a film of a blend of 85 parts of a linear ethylene/octene-1 copolymer having a density of about 0.920 g/cm³ and a melt index of about 0.75 dg/min and 15 parts of a high pressure polyethylene having a density of about 0.918 g/cm³ and a melt index of 8.5 dg/min; said interposed layer of polyethylene being of a thickness in the range of 5 to 110 microns and being formed from polyethylene having a density of at least $0.93~\mathrm{g/cm^3}$ and a melt index of less than 5 dg/min; said multilayer film having a thickness of from 40 to 130 microns.

22. A process for making pouches filled with a flowable material, using a vertical form, fill and seal apparatus, in which process each pouch is made from a flat web of film by forming a tubular film therefrom with a longitudinal seal and subsequently flattening the tubular film at a first position and transversely heat sealing said tubular film at the flattened position, filling the tubular film with predetermined quantity of flowable material above said first position, flattening the tubular film above the predetermined quantity of flowable material at a second position and transversely heat sealing said tubular film at the second position, the improvement comprising making the pouches from a flat web of film made from two layers of a sealant film and an interposed layer of polyethylene;

said sealant film being made from a composition comprising 10 to 100 parts by weight of a copolymer of ethylene and at least one C_4 - C_{10} alpha-olefin manufactured in a polymerization process using a single-site polymerization catalyst and from 0 to 90 parts by weight of at least one polymer selected from the group

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consisting of a linear copolymer of ethylene and at least one C_4 - C_{10} alpha-olefin having a density of from 0.900 to 0.930 g/cm³ and a melt index of from 0.3 to 2.0 dg/min, a high-pressure polyethylene having a density of from 0.916 to 0.930 g/cm³ and a melt index of from about 1 to 10 dg/min, and blends thereof; said interposed layer of polyethylene being of a thickness in the range of 5 to 20 microns and being formed from polyethylene having a density of at least 0.93 g/cm³ and a melt index of less than 5 dg/min; said film having a thickness of from 50 to 130 microns.

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23. A process for making pouches filled with a flowable material, using a vertical form, fill and seal apparatus, in which process each pouch is made from a flat web of film by forming a tubular film therefrom with a longitudinal seal and subsequently flattening the tubular film at a first position and transversely heat sealing said tubular film at the flattened position, filling the tubular film with predetermined quantity of flowable material above said first position, flattening the tubular film above the predetermined quantity of flowable material at a second position and transversely heat sealing said tubular film at the second position, the improvement comprising making the pouches from a flat web of film made from two layers of a sealant film and an interposed layer of polyethylene;

said sealant film being made from a composition comprising 10 to 100 parts by weight of a copolymer of ethylene and at least one C_4 - C_{10} alpha-olefin manufactured in a polymerization process using a single-site polymerization catalyst and from 0 to 90 parts by weight of at least one polymer selected from the group consisting of a linear copolymer of ethylene and at least one C_4 - C_{10} alpha-olefin having a density of from about 0.900 to 0.930 g/cm³ and a melt index of from 0.3 to 10.0 dg/min, a high-pressure polyethylene having a density of from 0.916 to 0.930 g/cm³ and a melt index of from about 1 to 10 dg/min, and blends thereof, said sealant film being selected on the basis of providing pouches with an M_{11} -test value that is lower, at the same film thickness, than that for pouches made with a film

of a blend of 85 parts of a linear ethylene/octene-1 copolymer having a density of about 0.920 g/cm³ and a melt index of about 0.75 dg/min and 15 parts of a high pressure polyethylene having a density of about 0.918 g/cm³ and a melt index of 8.5 dg/min.

24. The process of Claim 22 or Claim 23 in which the copolymer made with a single-site catalyst has a density in the range of 0.88-0.93 g/cm³; a stress exponent of less than 1.2; and a melt index of less than 5 dg/min.

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- 25. The process of any one of Claims 22-24 in which the copolymer made with a single-site catalyst is selected from ethylene/butene-1 copolymers, ethylene/hexene-1 copolymers, ethylene/octene-1 copolymers and ethylene/hexene-1/butene-1 copolymers.
- 26. The process of any one of Claims 22-24 in which the sealant film contains 20-100 parts of the copolymer manufactured with a single-site catalyst.
- 27. The process of any one of Claims 22-24 in which the sealant film contains 50-100 parts of the copolymer manufactured with a single-site catalyst.

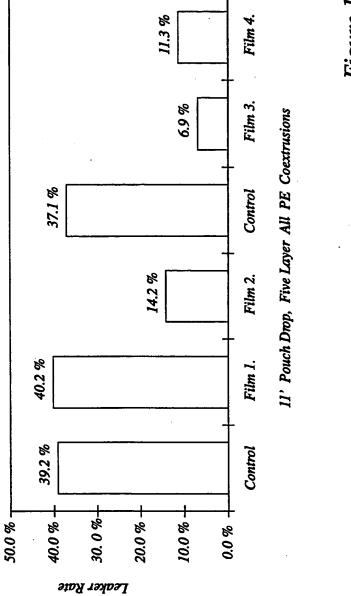


Figure 1.

INTERNATIONAL SEARCH REPORT

Intern. 1 Application No PCT/CA 95/00064

A CLAC	STEECH TROLL OF STEEL STEEL				
ÎPC 6	SIFICATION OF SUBJECT MATTER B32B27/32 C08L23/16 C09J12	3/16			
According	to International Patent Classification (IPC) or to both national cla	assification and IPC			
B. FIELD	S SEARCHED				
Minimum IPC 6	documentation searched (classification system followed by classifi B32B	cation symbols)			
Documents	ation searched other than minimum documentation to the extent th	at such documents are included in the fields	searched		
Electronic	data base consulted during the international search (name of data l	base and, where practical, search terms used))		
C. DOCUM	MENTS CONSIDERED TO BE RELEVANT				
Category *	Citation of document, with indication, where appropriate, of the	relevant passages	Relevant to claim No.		
X	WO,A,93 03093 (EXXON CHEMICAL PA	•	1-27		
	see page 1, line 37 - page 2, li see page 7, line 23 - page 11, l	ine 2 line 17			
	see page 13, line 36 - page 14,	line 15			
	see page 25, line 24 - page 26, see page 34, line 1 - page 36, l	line 51			
	page 34, Time 1 - page 30, 1	The Z			
X	WO,A,92 14784 (EXXON CHEMICAL PAINC.) 3 September 1992		1-27		
	see page 3, line 22 - page 7, li see page 19, line 8 - line 30 see page 20, line 1 - line 10	ne 11			
	see claims		-		
		,			
		-/			
		_ <u>_</u>			
χ Furth	er documents are listed in the continuation of box C.	Patent family members are listed in	n annex.		
* Special cat	egories of cited documents:	T later document published after the inte	rnational filing date		
conside	document defining the general state of the art which is not considered to be of particular relevance or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention				
E" earlier d' filing d	locument but published on or after the international ate	"X" document of particular relevance; the cannot be considered novel or cannot	claimed invention		
MDICD 1	nt which may throw doubts on priority claim(s) or s cited to establish the publication date of another	involve an inventive step when the do "Y" document of particular relevance; the	cument is taken alone		
'O' docume	or other special reason (as specified) nt referring to an oral disclosure, use, exhibition or	cannot be considered to involve an in- document is combined with one or me	ventive step when the		
other m P document	nt published prior to the international filing date but	ments, such combination being obvior in the art.	15 to a person skilled		
	an the priority date claimed	*A* document member of the same patent			
		Date of mailing of the international ser	иси героп		
2	May 1995	0 1. 06. 95			
Name and m	ailing address of the ISA	Authorized officer			
	European Patent Office, P.B. 5818 Patentiaan 2 NL - 2280 HV Rijswijk				
	Tel. (+31-70) 340-2040, Tm. 31 651 epo nl, Fax: (+31-70) 340-3016	De Jonge, S			

INTERNATIONAL SEARCH REPORT

Intern al Application No PCT/CA 95/00064

C.(Continue	DOCUMENTS CONSIDERED TO BE RELEVANT	
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
х	EP,A,O 580 377 (SUMITOMO CHEMICAL CO) 26 January 1994 see page 2, line 1 - page 3, line 30 see page 4, line 34 - line 38	1,2, 10-19
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